Doubly Focusing Crystal Analyzer for X-ray Fluorescence Holography

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ABSTRACT

Our goal was to prepare an X-ray Fluorescence Holography (XFH) experiment at ESRF, in which the holographic information is $\sim$ 0.3% of the overall isotropic fluorescent radiation. This requires a very pure fluorescent signal and the highest possible count rate. Therefore, we designed a focusing analyser system with large solid angle acceptance. It consists of a sagittally bent pyrolithic graphite crystals with large solid angle acceptance used in the parafocusing mode for the meridional plane. We successfully applied it to collect a large fraction of isotropically emitted Fe, Ni and Pd K_α fluorescent photons with the [002] and [006] graphite reflections, respectively. After characterising the phase space efficiency (angular acceptance, focal spot shape) of a 200 mm long, doubly focusing analyser, we developed a full circular one (220 mm diameter) to increase the solid angle acceptance to $\sim 2\ 10^{-2}$ sr, in view of an XFH experiment on MBE grown thin films. This analyser can be used more generally at K and L edges for atoms with Z $\geq$ 20.

Keywords: Mosaic crystal, X-ray Optics, Sagittal Focusing

1. INTRODUCTION

X ray Fluorescence Holography (XFH) is a developing technique that was first demonstrated by Faigel et al. in 1996.[1] Based on the same concept as photo-electron diffraction, but with X-ray fluorescent photons, the technique consists in measuring the interference between the fluorescent radiation directly emitted by the excited atoms and the same radiation scattered by neighbour atoms. In practice one has to measure the fluorescent intensity as a function of the emitted direction on (as much as possible of) the full solid angle. This method, where the fluorescent atoms inside the sample act as a source and the intensity is measured in the far field, is called normal X-ray fluorescence holography. For the demonstration experiment, done using a conventional X-ray generator on a bulk sample with a Ge solid state detector, one month of measurement was necessary. Soon after the first demonstration experiment, T. Gog et al. proposed a different approach to the X-ray holography (called here inverse XFH), by applying the optical reciprocity principle of exchanging source and detectors.[2] In that case, the fluorescent atom becomes the detector for the interference inside the sample of the direct incident beam and its scattering by the neighbour atoms. In practice, one has to measure the total fluorescent yield emitted as a function of the direction of the incident X-ray beam. If the detector system does not collect the whole solid angle it is necessary to move it together with the sample. We will hereafter only discuss the inverse holography mode, for which the detector has to collect the largest possible solid angle. The transposition to a high brilliance synchrotron radiation source requires a different approach for the detector system. Since the holographic signal is a few 10^{-3}, a typical hologram requires about 1 milion counts per measured point. Its modulation being a few degrees wide, the number of sampling points should be around 10^4 over the 4π solid angle. Therefore the counting rate should be in the range 0.1 to 1 MHz in order to obtain a complete data set within one day. At such a high counting rate, a conventional solid state detector cannot be used, and we have opted for an avalanche detector (APD). However the APDs, or diodes in current mode, present bad or no energy resolution, and unwanted radiations (Bragg diffraction peaks, inelastic scattering) cannot be discriminated. Therefore we decided to use in front of the detector an analyser crystal. In order to collect the maximum solid angle, we used the principle of a sagittally curved pyrolithic graphite analyser crystal (XTA) working in parafocusing condition (1:1 geometry). Based on a first prototype,[3] we developed and characterised larger XTAs

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suited to be optically coupled to the APD detector ($8 \times 8\ mm^2$ active area) . Due to the very small signal to be measured in different conditions, the stability of the setup and the optical quality are crucial. In this paper, we study the optical characteristics of these XTAs, such as the image spot at and around the focal distance, the reflectivity of different parts of the XTA etc. These informations can be used to study symmetry properties that allow to filter out systematic errors in the holographic measurement.

2. DESCRIPTION OF THE XTA

The first idea of the XTA consisted in an enlargement of the prototype, while keeping a variable radius of curvature. We fabricated several crystals by gluing several pieces (30×35 mm^2) of pyrolithic graphite with 1^0 degree mosaic onto 200×35 mm Cu-Be plates. Additional parallel grooves were cut every 2 mm to accomodate the relatively high curvature of the graphite (graphite thickness of 0.3 mm). However, tests of the analyser-detector assembly showed alignment difficulties. In addition, during the holographic scans, even a single XTA with variable radius of curvature was not stable enough when the detection arm moves. Hence we opted for a XTA having a fixed radius. In its first version, we glued the XTA inside a half-moon shaped hollow support. Then, in order to further increase the angular acceptance of the analyser, we designed at a later stage a second version consisting in a fully circular mount. Using the smallest possible radius of 110 mm, the detection system (XTA+APD) is compact enough - less than one meter for Fe K_α radiation and other 3d elements - to be moved by a diffractometer. Moreover, thicker graphite plates of 0.5 mm allow good reflectivity also at higher energies, e.g. the fluorescence from the 4d elements using the third harmonic reflection [006]. A photograph of the XTA (version 2) Fig. 1 shows its mounting assembly in 4 quadrants. Adjustments screws were foreseen, but in practice revealed to be unnecessary. The setup for holographic measurements (c.f. 4) is shown in Fig. 1b.

3. CHARACTERIZATION

We have characterised the first and second versions of our XTA at the ESRF optics beamline (BM05), with the setup shown in fig. 2.

3.1. XTA version 1

A nickel sample is excited by the white beam from the bending magnet source. the isotropic fluorescence is partially reflected by the XTA onto the focal spot. The detector equipped with a pair of slits is placed at the image focal distance. It analyses the shape of the focal spot ($5 \times 5\,\mathrm{mm}^2$)in the two transverse directions using a rotation around the XTA and a vertical translation. In addition, by changing the XTA - detector distance, one can follow the corresponding evolution of the spot and determine its optimum location. We have also characterised the angular efficiency of the XTA 2b; this was performed with the detector put on the focal spot, slits wide open, by moving another vertical slit located near the XTA. The Ni K_α and K_β fluorescence were both well separated by the XTA, giving an idea of the energy resolution (better than 5 %). The efficiency of the reflection was checked to be about 30 % at 7 keV, in agreement with previous results.[3]

3.2. XTA version 2

The second XTA was characterized in a similar way on the Optics Beamline, with the difference that the white beam from the bending magnet was monochromatized by the same graphite used for the XTA, and a Ge crystal used as the fluorescent sample at 10 keV. Additional characterization were performed during the holography experiment on an undulator source equipped with a Si(111) monochromator. The mosaicity was measured on a flat graphite piece to be fwhm 0.8 degree. One calculates straightforwardly a solid angle of acceptance of around $2\,10^{-2}$ sr. In the first instance, the XTA was mounted too high with respect to the beam, and without orientational adjustment. As a result, only the upper half circle of the XTA was characterised. By switching the position of the 4 parts, we could however check that the reflecting properties of the four quadrants were very similar. When placed on the diffractometer to measure a hologram, additional photos were taken to align the beam with Fe fluorescence at 6.4 keV. On Fig. 4a one can see the stronger K_α radiation in the focus, and K_β somewhat out of focus appearing as a fainter annular spot.

The angular efficiency was measured by placing a disk with polar slits just before the XTA. One can thus observe on Fig. 4b intensity variations due to the individual graphite pieces as well as the parallel grooves. The main parameters of the two versions of the XTA are gathered on Table 1.

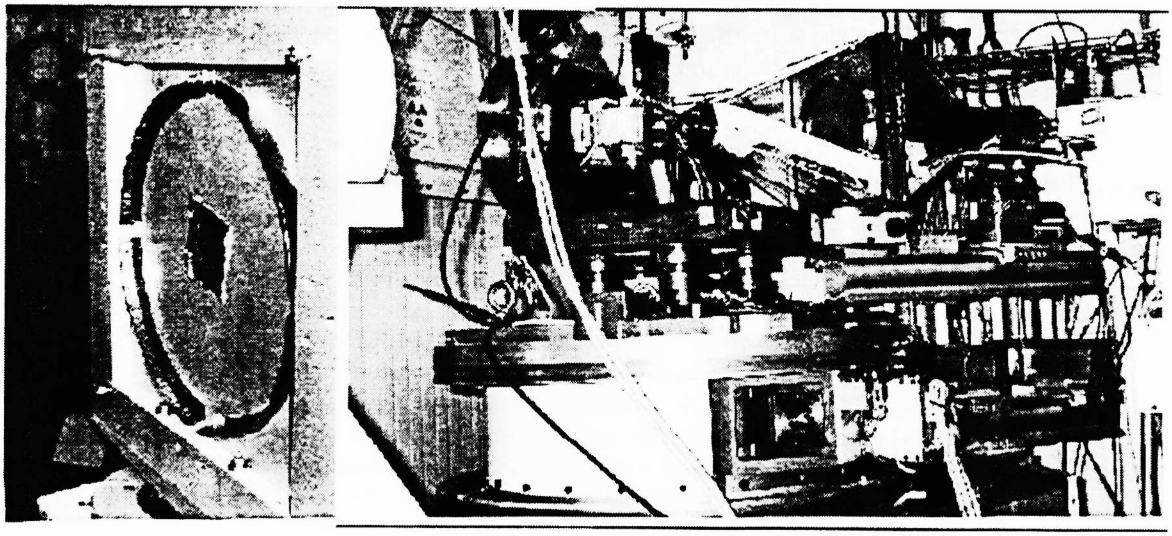

Figure 1. a) Photograph of the circular XTA (version2). b) Holography setup on ID32 beamline at ESRF, including the XTA in a He box

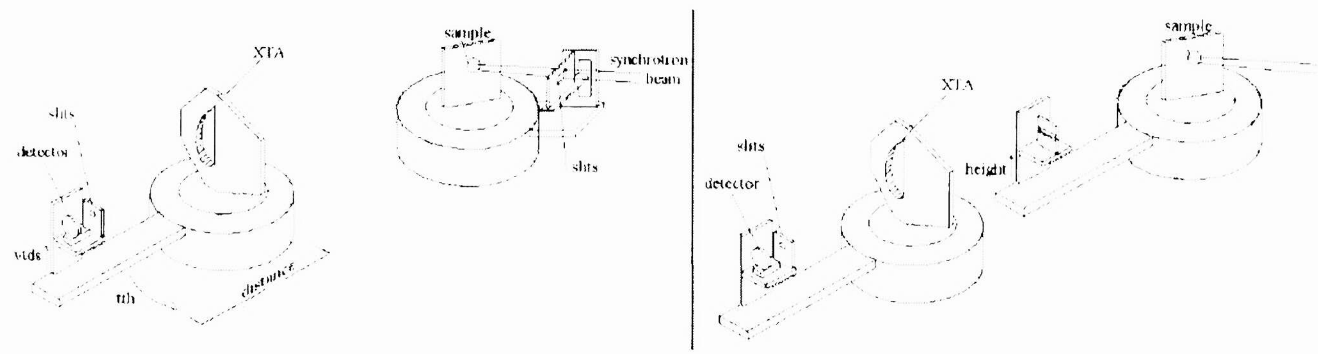

Figure 2. a) Setup used to image the focal spot from XTA (version 1). b) Modified setup (additional slits near the XTA for testing the reflectivity of the different portions of the XTA).

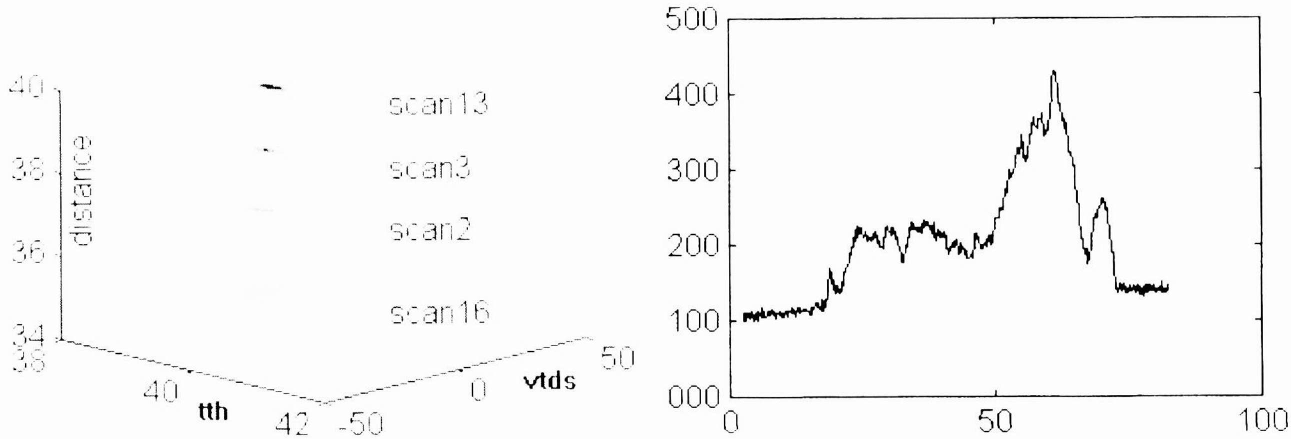

Figure 3. a) Measurement of the intensity at and near the focal spot. b) Measurement of the reflectivity of different parts of the analyser

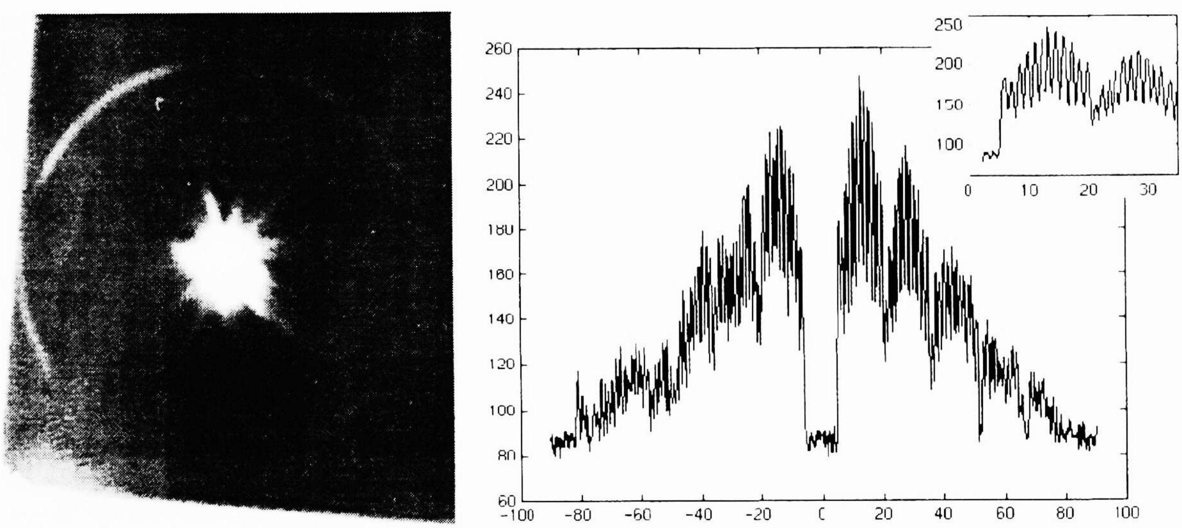

Figure 4. a) Image of the focal spot (fwhm $4 \times 4\ mm^2$) from XTA version 2 b) Polar slit analysis of the XTA reflectivity of the upper half part; the smooth decrease is due to a slight misalignment, while the insert clearly shows the two types of modulations (graphite pieces, parallel grooves)

Table 1. Main parameters of the two versions of the XTAs

	XTA1	XTA2
Material	Pyrolithic Graphite	Pyrolithic Graphite
Source	Advanced Ceramics	Optigraph
Overall dimension	$35\ mm \times 200\ mm \times 200\ \mu m$	$33\ mm \times 670\ mm \times 500\ \mu m$
D-spacing	3.355 Å	3.355 Å
Radius of curvature	130 mm	110 mm
Acceptance *	$5\,10^{-}3$ sr	$2\,10^{-2}$ sr
Spot size	$2 \times 4\, mm^2$	$4 \times 4\, mm^2$
focal distance $f = R_s/\sin(\theta_B)$	35.2 mm/keV	29.8 mm/keV
Parafocusing	1:1 geometry	1:1 geometry

4. HOLOGRAPHIC MEASUREMENTS

We have effectively used these XTA graphite analysers in few holographic measurements. However the first scanning technique used was too sensitive to mechanical stability and beam motion, leading to a variable overlap between the illuminated spot on the fluorescent sample and the optical acceptance of the XTA-APD system. This produced low frequency intensity variations that masked the holographic information. Nevertheless the higher angular frequency showed the full Kossel lines and X-ray standing waves patterns, respectively, when performing direct and inverse holography methods.[4] Since most of the difficulty arose from the motion the detector arm, we use at present a new scanning mode in which the azimuth scans are done at a fixed detector position. In this case the inverse holography is mixed with some direct contribution and we developed an algorithm to correct it. Holograms have been successfully obtained on model single crystals of CoO and NiO(111).[5]

The version 2 of the XTA (full circle) was used only very recently in a measurement on the ESRF ID32 beamline. Without filters we estimate 10^8 collected fluorescent photons of 6.4 keV, with more than 90 % efficiency of the APD, from a bulk Fe sample excited with 15 keV undulator radiation monochromatised by Si(111) ($\sim 10^{12}$ photons). In principle one should thus be able to record such an hologram in $\sim 10^2 - 10^3$ seconds. Another application for which the largest solid angle is mandatory is the study of epitaxial thin films. We have been able to apply, for the first time to our knowledge, the XFH method to MBE grown FePd systems, 70 nm thick.

5. CONCLUSION

We have developed and tested a wide angle doubly focussing XTA for X-ray Fluorescence Holography. Through a continuous interplay between optical characterisation and holographic measurements, we have been able to adapt this new method on a synchrotron source.

Acknowledgements

This work is an extension of the results from a collaboration on X-ray Fluorescence Holography and its developpement with M. Tegze and G. Faigel. We acknowledge the technical support from ID32 Beamline. We would like to thank M. Navizet for the first XTA. This work was supported by EC grant Contract ERBFMBICT961366 (S. Marchesini).

REFERENCES

1. M. Tegze, G. Faigel, , Nature **380**, (1996) 49.
2. T. Gog, P. M. Len, G. Matterlik, D. Bahr, C. S. Fadley, C. Sanchez-Hanke, Phys. Rev. Lett. **76**, (1996) 3132
3. A. K. Freund, A. Munkholm, S. Brennan 68/SPIE **2856** (1996)
4. S. Marchesini et al., Solid State Comm **105**(15), 685-687, 1998.
5. S. Marchesini, G. Faigel, M Tegze, M. Belakhovsky, *A comparative study of laboratory and synchrotron x-ray holography experiment* (submitted).